

given in Gray and Mathew's treatise and of these and a few other orders in Jahnke and Emdes' "Functionentafeln"—the writer does not recollect seeing the march of the functions illustrated for integral values of the parameter  $n$ .

In concluding this short note the author wishes to thank Dr. A. R. Forsyth, F.R.S., for the interest which he has shown in the work, which was carried out at the City and Guilds (Engineering) College.

XVIII. *An Investigation of Radium Luminous Compounds.*  
By CLIFFORD C. PATERSON, J. W. T. WALSH and W. F. HIGGINS.

RECEIVED APRIL 27, 1917.

*Synopsis.*

1. Introduction.
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1. *Introduction.*

In the present Paper is given an account of work carried out at the National Physical Laboratory on Radium Luminous Compounds. The investigations originated in a request by the Admiralty for information and assistance in connection with the use of this material for the dials of large numbers of instruments used by the Navy, and it is by permission of Capt. Creagh-Osborne, Admiralty Superintendent of Compasses, that there are included in this publication details of work which has been carried out at his request. The Compass Department of the Admiralty began the employment of radium luminous compound as far back as 1914, and was the first in the field to adopt it on an extensive scale. The use of separate lighting arrangements for instruments which require to be read at night is thus obviated.

The authors wish to say that there has been no leisure or opportunity to carry out a thorough investigation of the whole subject. Such an investigation would have included inquiry into the origin and characteristics of the various zinc sulphides used and of the effect of impurities on the luminosity of the compound, together with the effect of different methods of infusing into the zinc sulphide the very small quantities of radium which are used. These and similar problems have had to yield place to the necessity for fixing and applying proper



SDMS DocID 2053183

standards of luminosity both to the paint as manufactured, and to the instrument dials after the application of the luminous compound.

The standard of luminosity for the paint (generally known as the N.P.L. Standard) was fixed in absolute measure in December, 1915, and that for the dials themselves shortly afterwards, and many thousands of luminous instrument dials have since been measured for conformity with the standards laid down.

The subject is a comparatively new one, for the value of radium luminous compound has only been recently recognised in connection with commercial apparatus. Photometric measurements of the brilliancy of zinc sulphide under the action of the radiations from radium products were made by Marsden in 1910,\* who directed the radiation from a tube of emanation on to a zinc sulphide screen and measured the luminosity produced. The luminosity was found always to decay at a more rapid rate than the emanation.

In a Paper in 1915,† Mr. H. Hughes, describing the Creagh-Osborne compass, dealt with the use and application of radium compound to prismatic compasses, and in a further Paper to the Optical Society in June, 1916, F. Harrison Glew‡ dealt with the general question of luminescence, and considered the practical uses to which radium luminous compound may be put, as well as the method of application of the compound to a surface in order to produce the maximum luminosity. Mr. Glew gave a further lecture on this subject to the Illuminating Engineering Society,§ and this was followed by notes on the behaviour and testing of luminous paint by Mr. J. S. Dow and Mr. A. Blok.

A description of the apparatus used at the Bureau of Standards for measuring the luminosity of radium luminous compound has been given by N. E. Dorsey.||

In the present Paper the ground covered by these authors will only be touched upon in so far as is necessary for the present purpose. It may be recalled, however, that radium

\* Roy. Soc. "Proc." LXXXIII, pp. 548-561, 1910. "Sci. Abst.," 76 A., 1911.

† "Improvements in Prismatic Compasses," H. A. Hughes, "Trans." Opt. Soc., Lond., Vol. XVI, p. 17.

‡ "Radium and Other Luminous Compounds," F. Harrison Glew, "Trans." Opt. Soc., Lond., Vol. XVI, p. 276.

§ "Illum. Eng.," p. 72, March, 1917.

|| "Washington Acad. Sci. J.," 7, pp. 1-6, Jan. 4, 1917.

luminous compound consists almost exclusively of specially prepared zinc sulphide, with which is mixed about four parts per 10,000 of radium bromide or its equivalent in gamma ray activity.

The luminosity of the resulting powder depends on the amount of radium present and on the quality of the zinc sulphide. The reason for the superiority of one type of zinc sulphide over another is at present obscure, but the responsiveness of different grades of zinc sulphide to radioactive excitation varies very greatly and offers a wide field for investigation. The zinc sulphide responds mainly to the  $\alpha$ -ray activity of the radium (see Glew, *loc. cit.*). As radium is bought, sold and measured by its  $\gamma$ -ray and not by its  $\alpha$ -ray activity, a constant ratio is assumed between the two, and all values of radium content given in this Paper are in terms of  $\gamma$ -ray activity.  $\alpha$  rays are emitted from the radium itself and from the emanation, but they are readily stopped by solids (including any organic medium used for binding). According to the so far generally recognised hypothesis the  $\alpha$ -particles impinging on the crystals of zinc sulphide excite certain active centres. After its bombardment each active centre becomes extinct and a gradual decay of the zinc sulphide therefore results. That this theory, in its simple form, fails exactly to represent the facts is shown later, and a modification of this theory is discussed in a second theoretical Paper by one of the authors.\* To apply the paint in practice the powder is mixed with a minimum possible quantity of varnish or other suitable binding material, and laid on either with a brush or by filling in the figuring which has been engraved into the surface of the object to be painted.

## 2. Measurements of Luminosity.

The measurement of the luminosity of radium paint is rendered difficult by two considerations—(i.) the green hue of the light emitted, (ii.) the very low brightness to be measured. It is desirable at the outset to make determinations of luminosity in absolute rather than relative measure, and comparisons have therefore to be made with the ordinary photometric standards.

(i.) The very green hue of the light given by radium compound necessitates the use of a green filter when photometering

\* "The Theory of Decay in Radioactive Luminous Compounds," J. W. T. Walsh.

it. Marsden examined the light spectroscopically, and found that the visual region lay between  $\lambda=5,920$  and  $\lambda=4,250$ , with a fairly sharp edge towards the red. It is found that Wratten Filter "Minus Red," used in conjunction with a tungsten filament lamp operating at about 1.5 watts per candle, gives an approximate colour match with the samples of radium paint ordinarily used. A still better match is obtained by the use of a sample of glass made by Messrs. Chance Bros., known as "B.O.T. Light Green," and corresponding with the lightest shade of green glass permitted for ships' starboard navigation lights. The expression of the luminosity in absolute measure entails the determination of the transmission coefficient of the filter. This has been carried out at the Laboratory by a number of observers, so that the result may correspond as nearly as possible with that which would be given by the "average" eye.

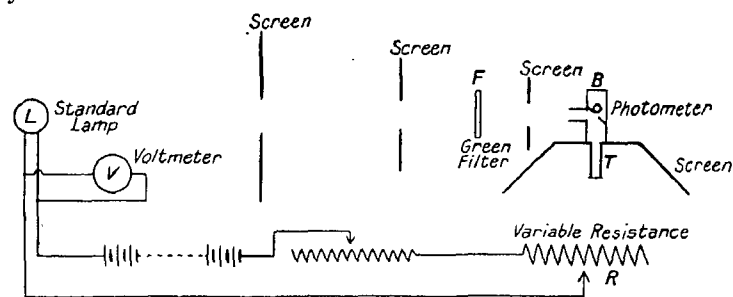


FIG. 1.—DIAGRAM OF APPARATUS FOR MEASURING THE BRIGHTNESS OF RADIUM LUMINOUS COMPOUND.

(ii.) The degree of brightness required to be measured is of the order of 10 microcandles per square centimetre, or 0.03 equivalent foot-candles.\*

This is about one-hundredth of the brightness of a newspaper satisfactorily illuminated for comfortable reading by artificial light. The brightness is of the order of that of white paper illuminated by the light of the full moon. It will be seen that this degree of illumination is one at which the Purkinje effect is operative, so that the transmission coefficient of the green filter must be determined at an illumination corresponding with the brightness of the paint with which it is to be used,

\* This is a convenient unit of brightness, and represents a brightness equal to that of a perfectly diffusing surface with a coefficient of reflection of 100 per cent., when illuminated to an intensity of 1 ft.-candle (i.e., by a source of 1 candle placed 1 ft. away from the surface in the direction of its normal).

and it must be ensured that the surfaces to be compared are really in juxtaposition without any intervening space to separate their images on the retina.

The following description and diagram (Fig. 1) show a form of apparatus which 18 months use at the Laboratory has shown to be eminently satisfactory for this class of measurement.

L is a tungsten filament lamp mounted on the carriage of a 30 ft. long photometer bench. It is fed from a storage battery, the pressure at the lamp terminals being controlled by a resistance, R, which is to the hand of the observer. The value of the pressure applied to the lamp terminals is indicated by a voltmeter, V. F is the special green filter. It reduces the hue of the light given by L to that of the radium compound. B is a small brass box, 120 mm. long by 37 mm. square, and shown in more detailed plan in Fig. 2.\*

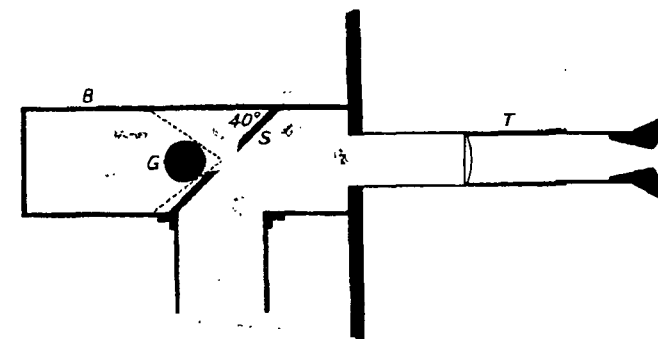


FIG. 2.—DETAIL OF BOX CONTAINING THE COMPARISON SURFACE AND TUBE OF COMPOUND.

It contains S, a sheet of matt white celluloid, set at an angle of 50 deg. to the axis, and having at its centre an oblong aperture, 4.5 mm. high by 7 mm. broad. The relative dimensions are such that when the screen is looked at through the telescope the aperture has the appearance of a small square with 4.5 mm. sides. This aperture is cut with a sharp bevel, so as to reduce to the smallest possible dimensions the black edge between the illuminated screen and the surface of the radium compound.

The radium compound is contained in a glass tube, G, and wedge-shaped guides are arranged inside the box to assist in placing it centrally behind the aperture in S, no matter what may be the diameter of the tube.

\* A very similar apparatus was described by Mr. Blok to the Illuminating Engineering Society, March, 1917.

The transmission coefficient of the filter  $F$  was determined from the mean results of five experienced photometric observers, each of whom matched the brightness of three separate samples of radium paint against the brightness of the screen  $S$ . The illumination of  $S$  was varied by moving the lamp  $L$  along the line  $LF$  until a match of intensity was obtained. This was done (i) with the filter  $F$  removed, and (ii) with the filter in place. The mean ratio of the illuminations was taken to represent, for the conditions under which the apparatus is used, the transmission coefficient of the filter for an "average" eye.

The values obtained for this coefficient by the different observers differed from the mean value by  $-17.2$ ,  $-6.4$ ,  $+4.3$ ,  $+8.5$ , and  $+10.8$  per cent. The mean value must not, therefore, be considered as accurate to nearer than 3.5 per cent. This inaccuracy in the absolute transmission coefficient may appear large at first sight, but a very much greater number of observers would be required to reduce it to 1 per cent.

It will be observed that the transmission coefficient was determined under conditions identical with those prevailing when the luminous compound itself is measured—i.e., the size of the field and its illumination are the same—so that those errors are avoided which result from the well-known retinal peculiarities which appear in the use of green light at low illuminations.\* The mean coefficient obtained was 2.15 per cent. This value was confirmed by means of a portable photometer, the illumination of a white card being measured directly (a) without the filter, (b) with the filter interposed.

The other constant to be determined was the coefficient of reflection of the white card  $S$  as viewed from the eyepiece  $T$ , with the light incident from  $L$  at an angle of  $40^\circ$ . This was determined by measuring the brightness of the card as viewed from the direction of  $T$  with a portable photometer, and comparing this with the brightness of another portion of the same piece of card when held normal to the direction of the light.

The coefficient obtained in this way embodied both the coefficient of reflection  $K_\theta$  of the card with the light incident at an angle  $\theta$ , and the reduction of illumination according to the cosine law. This combined coefficient  $K_\theta \cos \theta$  was found to be 68.2 per cent.

To obtain now the brightness of a sample of paint  $T$  from the

\* "Colour Phenomena in Photometry," J. S. Dow, "Proc." Phys. Soc., Vol. 20, 1906, p. 245.

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candle-power  $C$  of the lamp  $L$  at distance  $d$  when the brightnesses of  $S$  and  $T$  are equal, we have:—

$$B = \frac{C}{d^2} \times 0.215 \times 0.682.$$

There is, however, one more point yet to be considered. It will be clear that the transmission coefficient of the filter  $F$  will vary with the hue of the light traversing it, so that the value of the coefficient found above holds only for the light from a tungsten filament lamp operating at the one definite efficiency, at which the determination of its transmission coefficient was made. The result of this is that variation of illumination on  $S$  has to be obtained by change of position of  $L$  along the line  $LS$  rather than by change of voltage  $L$ . In actual use, however, when measuring the brightness of a sample of radium compound, it is found most convenient to fix  $L$  in such a position that an approximate match is obtained in  $S$  with a voltage on  $L$  of 95. Several settings are then made by means of the resistance  $R$  and as the extreme voltage variation on  $L$  thus obtained does not generally exceed 2 volts, it is safe to assume that over this range the variation in the hue of the light from  $L$  will not appreciably affect the value of the transmission coefficient of  $F$ . The candle-power variation with voltage of  $L$  has been determined accurately on the photometer bench, over the range from 92 to 98 volts, and from the voltage indicated by  $V$ , the distance  $LS$ , and the formula given above, the brightness of the paint in  $T$  is at once found.

*Absorption of Glass Tubes.*—The radium paint being in powder form, it is necessary to measure its brightness when enclosed in a glass tube. This is desirable for two reasons. Firstly, it is necessary to obtain a smooth surface of the powder in order that it shall be of uniform brightness, because any cavity in the surface shows a greatly enhanced brilliancy due to internal reflection. Secondly, it is necessary to avoid any possible loss of emanation.

From a series of experiments made when the apparatus was first set up, it was found that variations in the thicknesses of the glass containing tubes, up to at least 1.3 mm. makes no difference to the brightness of the paint viewed through them.

The colouration of the glass by the action of the radium, on the other hand, produces a progressively increasing absorption for which allowance has to be made in considering the

decay of luminosity of the compound.\* Lead glass gives a brown coloration and soda glass a purple one.

Measurements have been made of the amount of this effect in the case of six of the samples of compound studied, all of which were contained in soda glass tubes. The absorption was found by making alternate measurements of the luminosity of a sample of compound when contained (a) in the vessel under examination, (b) in a new vessel not previously exposed to  $\gamma$ -radiation. The results obtained are tabulated below and the value of luminosity given throughout the Paper are corrected on the assumption that the light absorption has followed a linear law of increase. As the whole correction does not in any case exceed 16 per cent. this assumption will not lead to any appreciable error in the final corrected values of luminosity.

TABLE I.

Sample.	Age of compound.	Total absorption due to coloration.
3	14½ months.	13 per cent.
6	17 "	8 "
7	17 "	9 "
8	11½ "	10 "
9	13 "	10 "
10	13 "	16 "

### 3. Measurement of Radium Content.

The general method of determination of the quantity of radium contained in a concentrated sample is by direct comparison with a standard consisting of a known amount of one of the salts of the element. The means by which this comparison is carried out is based on the ionisation produced by the  $\gamma$ -rays emitted from the samples considered.

Radium gives out three types of rays—namely, the  $\alpha$ ,  $\beta$  and  $\gamma$ -rays—and the ionisation produced by any of these could be made use of as a measure of the quantity of radium present. Two difficulties, however, arise in attempting to use either of the two former types of radiation. Firstly, in the case of radium in radioactive equilibrium, it is not possible to isolate these rays, so that the properties of the  $\alpha$  or  $\beta$ -rays alone are employed. The same trouble does not arise in the case of  $\gamma$ -rays as these are very penetrating and may consequently be freed from  $\alpha$  or  $\beta$  rays by the use of suitable screens of absorb-

\* Glew, *loc. cit.*, p. 287.

ing material.  $\alpha$ -rays are absorbed by about 8 cm. of air at atmospheric pressure or by very thin layers of solid matter, while the  $\beta$ -rays are completely stopped by about 1 cm. of lead. In practice it is found that a sheet of lead 5 mm. thick reduces the proportion of  $\beta$ -rays present to a negligible amount compared with the  $\gamma$ -rays. Consequently in the apparatus to be described later the ionisation chamber is covered by a lead disc 5 mm. thick, and the ionisation produced inside this space must therefore be solely due to the  $\gamma$ -rays. In the second place the intensity of all the rays changes with time until complete radioactive equilibrium is attained. Consequently the  $\alpha$  or  $\beta$ -ray activity of any sample of radium and its radioactive products will not be sufficiently constant for measurement purposes for some years. For the  $\gamma$ -rays, however, a constant intensity is reached in a comparatively short time (about 30 days suffices) as the products of disintegration beyond radium do not emit  $\gamma$ -rays (with the exception of some very soft rays, which in practice are completely absorbed before the ionisation chamber is reached). For purposes of measurement it is necessary to prevent the escape of the gaseous product—viz., radium emanation—and to this end the material must be sealed up in glass or metal tubes for a sufficient length of time before comparisons can be undertaken.

The  $\gamma$ -ray method is primarily intended for the estimation of high concentration, but may also be extended to estimation of the total quantity of radium in a sample of radium luminous compound if the necessary precautions as detailed later are taken.

The smallest equivalent quantity of radium bromide which can in this way be accurately measured is about 1 mgm., and the quantity of radium luminous compound on which the test is made should hence be from 3 to 5 grms.

The standard against which the samples of radium luminous compound are compared at the National Physical Laboratory, consists of a preparation of pure radium bromide, equivalent to 5.20 mgm. of hydrated radium bromide ( $\text{RaBr}_2 \cdot 2\text{H}_2\text{O}$ ). This is compared at intervals with the British Radium Standard, which was certified by the International Radium Standards Committee after comparison with the International Standard at Sèvres.

The apparatus employed is represented in Fig. 3. It consists of a massive wooden bench along which a stand, A, can be moved. The position of this stand with respect to the

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ionisation chamber is indicated by a pointer and scale. The samples under test are placed in turn in the V-support on the stand *A*, and this is so arranged as to bring the sample approximately to the level of the centre of the ionisation chamber.

The ionisation chamber *C* consists of a lead cylinder of internal diameter 140 mm. and depth 94 mm. The thickness of the walls is 6 mm. The front of this chamber consists of a lead plate of 5 mm. thickness; this is held in close contact with the walls of the chamber by suitable clamping screws. Alternative lead plates of different thicknesses are available as desired. The central insulated disc is supported from the back

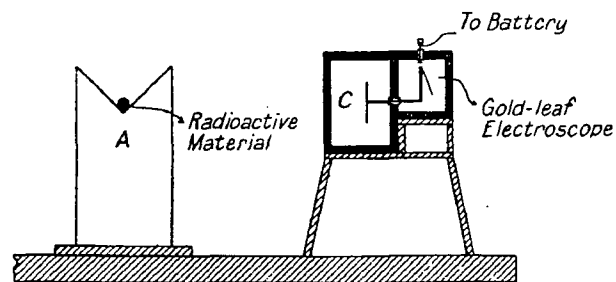


FIG. 3.—DIAGRAM OF APPARATUS FOR MEASURING RADIUM CONTENT BY THE  $\gamma$ -RAY METHOD.

of the chamber by means of a brass wire passing through an amberite\* bush.

The gold leaf electroscope by which the ionisation current is measured is attached to the ionisation chamber. The gold leaf is fastened on to a vertical projection from the brass supporting wire of the disc above referred to, and is surrounded by a square built up box of thick lead plates. Windows are provided at each side of this box by which the gold leaf may be illuminated and viewed. These windows are of lead glass, and are screened by lead shields to prevent as far as possible the entrance of stray and scattered  $\gamma$ -radiation. The insulated system, consisting of the disc in the ionisation chamber, the gold leaf and the support are positively charged by means of a

\* This material is composed of highly compressed amber chips or dust, and provides an excellent insulator, which may be turned to any desired shape. Care must be exercised in doing this as the material is somewhat brittle. It is not hygroscopic, nor does it show any appreciable soakage effect.

charging key carried on the top of the electroscope. On pressing down the plunger of this, contact is made with a battery of accumulators of the required voltage (about 200 volts). The case and all metal parts of the apparatus other than the central insulated system are earthed, as is also the negative pole of the battery.

The motion of the gold leaf is viewed through a reading microscope magnifying about 10 diameters, and provided with a scale in the eyepiece. This instrument is held rigidly by means of an adjustable stand, and when once put into position is not moved throughout the complete series of readings.

The readings are always taken over a definite portion of the eyepiece scale, the time being determined by means of a stop-watch for the gold leaf to move over this distance. The procedure generally adopted in making a comparison is as follows: The natural leak of the system is first determined. This is done by charging up the gold leaf and noting the time taken by it in moving over the measured distance, all radium being removed from the neighbourhood of the measuring instruments. The radium standard is then placed on the stand *A*, which is moved to some definite position along the test bench according to the strength of the sample to be measured. For small quantities the stand is brought relatively near the ionisation chamber, while for larger amounts the distance between the two is increased. The rate of leak is then determined for the standard sample, the mean of a number of observations being taken. The standard is then replaced by the material under test without the relative positions of any parts of the apparatus being changed and the rate of leak is again determined. Further readings of the standard are then taken, which should agree with those first obtained.

The comparison is then repeated for other distances between the stand *A* and the ionisation chamber *C*. Finally, the natural leak is taken at the end of the series of observations. It must be noted that all radium preparations except that actually on the test bench, must be removed as far as convenient from the  $\gamma$ -ray apparatus, otherwise the natural leak will be large, and the corresponding accuracy of the comparisons will be greatly reduced.

The correction for the natural leak is made thus: If  $T_0$  is the time taken for the leaf to move over  $n$  divisions when the sample of radium is under observation and  $T_n$  the number when it is

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absent (i.e., the natural leak) then the corrected value of the time ( $T'$ ) will be given by—

$$T = \frac{n}{T_0} - \frac{n}{T_n},$$

i.e.,

$$T = \frac{T_0 T_n}{T_n - T_0}.$$

The corrected value of the time is thus obtained for the sample under test and for the standard. The ratio of these times then gives the ratio of the quantities of radium present in the standard and the sample under test, thus:—

$$\frac{\text{Weight of radium in sample}}{\text{Weight of radium in standard}} = \frac{\text{Time for standard.}}{\text{Time for sample.}}$$

*Correction for Absorption.*—From the above description it will be clear that in determining the radium content of a sample of luminous compound by measurement of its  $\gamma$ -ray activity, allowance must be made for the absorption of the  $\gamma$ -rays by the zinc sulphide. If, first, we suppose that the whole of the radium salt is concentrated along the axis of a cylindrical vessel containing the compound, it is known that the intensity of the  $\gamma$ -ray activity in any direction perpendicular to the axis (and at a distance from it sufficient to make the length of the vessel negligible in comparison with this distance) will be given by  $I' = Ie^{-\lambda a}$ , where  $I'$  is the intensity which would be found in the absence of the zinc sulphide,  $a$  is the radius of the vessel, and  $\lambda$  is the absorption coefficient of the zinc sulphide.

In actual practice, however, the radium salt is not concentrated at the axis, but is uniformly distributed over the cross-section of the cylinder, so that the above formula does not accurately apply, although *a priori* reasoning from the known value of  $\lambda$  shows that for vessels of small diameter this formula is sufficiently accurate for all practical purposes. When the diameter reaches 20 mm. or 30 mm., however, it becomes necessary to investigate the effect of uniform distribution more thoroughly, and this may be done in one of two ways—(a) approximately, by first treating the simple case of a vessel of square section and then regarding the cylindrical vessel under consideration as made up of any convenient number of equal square-section vessels, laid alongside one another; or (b)

rigidly by obtaining a convergent series with constant coefficients which will give at once the required value of intensity, from a knowledge of the value of  $\lambda$ . Methods (a) and (b) are described in greater detail in Appendix A.

The value of  $\lambda$  has been determined experimentally by mixing a measured quantity of radium salt with an amount of zinc sulphide sufficient to produce radium luminous compound of standard specification, and then determining the  $\gamma$ -ray activity of the resulting mixture. The contents of a tube of radium salt equivalent to 11.4 mgm. of hydrated radium bromide, as determined by a  $\gamma$ -ray test were mixed, without loss, with 28.5 grms. of active zinc sulphide, and the mixture was enclosed in a glass bottle of known dimensions. The  $\gamma$ -ray activity was measured at intervals until a constant value was obtained. This was equivalent to 8.2 mgm. of hydrated radium bromide, and from this result a value for  $\lambda$  is calculated from the formula on page 226. This value is  $\lambda = 0.03 \text{ mm}^{-1}$ .

A confirmatory test was made by the use of a second sample consisting of 30 grams of radium luminous compound, which was measured in bulk, and then was subdivided into a series of six smaller quantities enclosed in glass tubes of various diameters.

The  $\gamma$ -ray activity of each of these was determined and was corrected for absorption in accordance with the value obtained in the first test. The radium content per gram. was found to be the same, to within an accuracy of 2 per cent., for each tube and for the sample in bulk when this had been done.

An attempt was also made to obtain the absorption coefficient by surrounding a sealed tube of radium bromide with a known thickness of zinc sulphide as mentioned above. The value so obtained leads to a result of  $\lambda = 0.0092_5 \text{ mm}^{-1}$ , which is roughly one-third of the value obtained in the case of uniform distribution. No explanation of this discrepancy is at present available, but further work is proposed to investigate the matter. In the meantime the first value is assumed to hold, as the conditions under which it is obtained most nearly represent the conditions of actual test of samples, and the table following gives the factors by which the measured radium content must be multiplied in order to give the true content for different sizes of containing vessels.

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TABLE II.  
( $\lambda = 0.03 \text{ mm}^{-1}$ ).

Diameter of containing vessel.	True content Measured content
5 mm.	1.06
10 mm.	1.13
15 mm.	1.20
20 mm.	1.28
25 mm.	1.35
30 mm.	1.43

#### 4. Measurement of Luminosity of the Markings of Luminous Dials.

The chief commercial use of radium luminous compound lies in its application to instrument dials for rendering the figures and graduations on these visible at night without external illumination. The method by which this application may be made has been described by F. H. Glew (*loc. cit.*), and it was early found that the various methods employed by different makers of the dials gave very widely differing results. The luminosity of the figures, therefore, depends not only on the quality of the radium luminous compound employed, but also on the method of its application to the dial, so that the employment of a good quality of luminous compound is not a necessary guarantee of a satisfactory dial.

It becomes necessary then, to test the actual brilliancy of the luminous markings, and the only satisfactory method of doing this is to compare the dial under test with another dial, the markings of which are of the same pattern and dimensions, and of a known brightness. Hence the first test employed was comparison with a radium painted dial of what was considered to be good luminosity. This was very soon abandoned, however, in favour of a more convenient and fundamental test. In the first place the possibility of a deterioration of luminosity was early recognised as one of the dangers of a radium-painted standard dial, while the large number of different patterns of dial to be tested would have necessitated a considerable collection of radium painted standards. These would necessarily have been at variance as regards brilliancy, since absolute uniformity is practically impossible of attainment, especially under the conditions prevailing at the time when the standards were first set up. A still more fatal drawback was the fact that beyond the statement that the test dial did or did not equal in brilliancy the standard dial, no value

could be assigned to the luminosity of its markings to serve as a guide for future work by the manufacturer.

In view of all these considerations a method was devised which has from the very first given complete satisfaction in working. This method is, in essence, the comparison of the test dial with a metal stencil cut to the same pattern and dimensions as the luminous markings of the dial, and having behind it a screen illuminated by light of the same hue as that given by the radium compound. The intensity of illumination of this screen is variable by means of a resistance in series with an electric lamp, and thus a knowledge of the variation of illumination of the screen with the current taken by the lamp gives at once the value of the brilliancy of the markings on the test dial. The actual apparatus is shown diagrammatically in Fig. 4, where a box encloses at one end an electric lamp,  $L$ , in

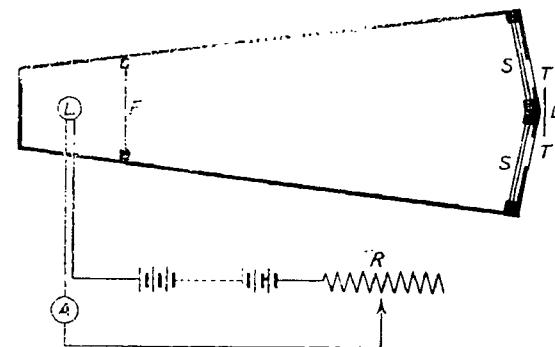


FIG. 4.—DIAGRAM OF APPARATUS FOR MEASURING THE BRIGHTNESS OF THE MARKINGS OF LUMINOUS DIALS.

circuit with a resistance,  $R$ , and precision ammeter  $A$ .  $R$  is to the hand of the observer.  $L$  is actually a 105-volt 60-watt metallic filament lamp operated, when giving standard brilliancy, at about 5 watts per candle. In front of  $L$  is a sheet of Wratten "Minus Red" filter  $F$ , in order that the hue of the light may match that of the radium compound. The green light then falls on two screens  $S, S$ , each consisting of two thicknesses of thin white paper with a space of 10 mm. between them. The two thicknesses of paper were found necessary to secure proper diffusion of the light and it was also found desirable to whiten the inside of the wooden strips separating the two layers of paper in order to avoid any decrease of luminosity at the edges. In front of the screens are placed two stencils  $T, T$ , each consisting of a sheet of thin brass or copper 0.3 mm. thick, framed in wood to fit the dimensions of



the screens ( $255 \times 275$  mm.). The figures and graduations are cut in brass on an engraving machine, so as to be identical with the luminous markings of the dials under examination. Between the stencils is fixed the dial under test *D*. It was originally intended that the test dial should be placed above the stencils so that the three dials under comparison should be one at each apex of an equilateral triangle. This arrangement was abandoned, however, as it was found during the course of the initial experiments that different luminosities were assigned to a dial according as it was placed above, below, to the left or to the right of the dial with which it was being compared. This effect is apparently one of personal error depending on the distribution of sensitivity to green light over the retina of the observer. In different observers the effect was found to vary in amount, and also to change sign. In these circumstances it was found best to give the dial an intermediate position between two comparison dials of equal luminosity. In making

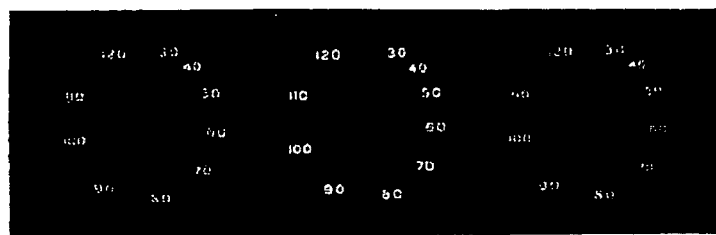


FIG. 5.—PHOTOGRAPH SHOWING THE APPEARANCE OF A LUMINOUS DIAL AND ITS COMPARISON STENCILS IN THE DIAL-TESTING APPARATUS.

a test of a luminous painted dial by this method the dial, as has been said, is placed between the two stencils of similar pattern, while the operator, seated with his eye on a level with and about 3 ft. from the dial varies the brightness of the comparison stencils by means of the resistance *R*. As soon as he is satisfied that the three dials are of equal luminosity he notes the current indicated by the ammeter *A*, and from a table finds the percentage of standard luminosity of the dial under test.

This test is, of course, performed in a dark room, and it is necessary for the observer to remain sufficiently long in the dark for his eyes to become "dark-adapted." The appearance of the test dial and its corresponding stencils is shown in the photograph (Fig. 5). It may be here noted that the luminosity of the markings on standard dials is 0.0075 equivalent

foot-candles (2.7 microcandles per square centimetre), and that to obtain the above photograph with an Imperial Standard rapid plate an exposure of  $2\frac{1}{2}$  hours was required.

An examination of the negative shows that, although there was a luminosity match between the three dials, the light from the centre one, painted with luminous compound, was more actinic.

##### 5. *Investigation of the Best Dimensions for Luminous Markings to Secure Maximum Legibility.*

In addition to the question of luminosity of the markings of a radium painted dial, there is also the consideration of legibility, which will depend on the form and size of the markings as well as on their luminosity. An investigation was undertaken at the request of the Admiralty to determine the influence of these factors.

It is obvious that if the luminous lines or markings of a card are reduced in breadth too much, the letters will become illegible, because the total amount of light has become so small.

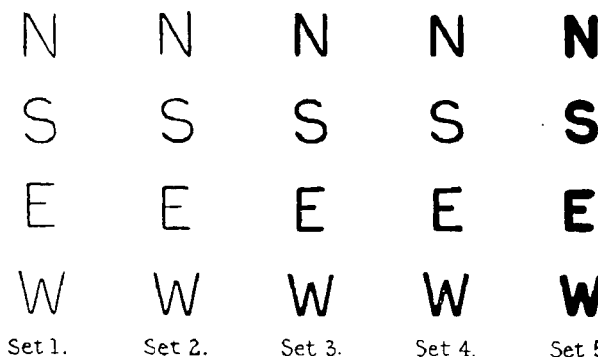


FIG. 6.—THE TYPES OF FIGURES USED TO INVESTIGATE THE RELATION BETWEEN DIMENSIONS AND LEGIBILITY.

Conversely, if the lines are increased in width beyond a certain point, in spite of the large amount of light emitted, the letters will lose legibility and appear merely as blotches of light. Between these two extremes there must lie a region where the legibility is a maximum. To determine this optimum width of line, observations were made on a series of letter stencils in which the letters were all 16 mm. high, but in which the width of line varied from 0.5 mm. to 3.0 mm. The letters are shown in Fig. 6. The first set with the thinnest line was used as a standard and the other sets were compared

with it. The specific luminosity of each set of letters was independently variable, and was so adjusted that the *total* light emitted was the same for each set. This adjustment was made by reducing the brightness of the broader letter by an appropriate number of thicknesses of fine black tulle. Thus, any two sets of letters when compared from a distance appeared equally bright, and it is of interest to note that under these conditions the difference in the width of the lines could not be detected.

If, however, instead of comparing the letter from the point of view of brightness, the observer made a judgment of the relative legibility, a difference was in some cases noticeable. The set having the broadest lines (set 5) compared with the thinnest (set 1) was clearly less legible. The falling off in legibility was not, however, so noticeable between sets 1 and 4, and was inappreciable between sets 1 and 3.

In order to get a quantitative measurement of the falling off of legibility of sets 5 and 4, their *luminosity* was increased until the *legibility* equalled that of set 1. Equality resulted when set 5 was increased about 40 per cent. and set 4 about 20 per cent. The results are summarised as follows :—

TABLE III.

—	Width of line.	Ratio of width of line to height of letter.	Legibility.
Set 1 .....	0.5 mm.	1/32	} All equally legible for equal total luminosity.
Set 2 .....	1.0 mm.	1/16	
Set 3 .....	1.5 mm.	1/11	
Set 4 .....	2.0 mm.	1/8	Fall of legibility = 20 p.c.
Set 5 .....	3.0 mm.	1/6	Fall of legibility = 40 p.c.

It will thus be seen that for a certain specific luminosity, set 5 will not appear very much more legible than set 3, because although set 5 is twice as bright, it suffers a falling off in legibility of about 40 per cent. An experimental trial was made to test these deductions and was found to confirm the results stated.

It was concluded from these experiments that the proportion of width of line to height of letter for maximum legibility should be about 1 to 8 (set 4), and should not much exceed 1 to 7, at any rate for letters and figures in which the height exceeds about 5 mm. Below this limit experience suggests that the breadth of line may be usefully increased to one-sixth of the height of the figure.

## 6. Decay of Luminosity.

Glew points out \* that the luminosity of radium compounds falls off with time. The radium itself decays to half its value in about 2,000 years, and it has sometimes been assumed that radium paint will retain its luminosity for a similar period. This is not, of course, the case, as Marsden showed in his experiments on zinc sulphide, willemite and barium platino-cyanide. His source of radiation, however, being radium emanation, was itself decaying at a comparatively rapid rate,

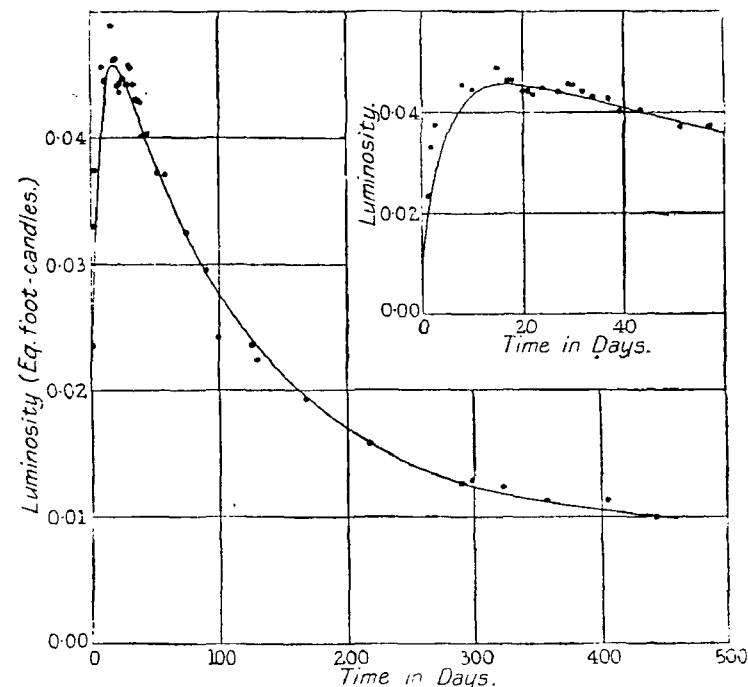


FIG. 7.—DIAGRAM SHOWING THE VARIATION THROUGHOUT LIFE OF THE LUMINOSITY OF SAMPLE 3 OF RADIUM LUMINOUS COMPOUND. INSET, THE EARLY PORTION OF THE LIFE ON AN ENLARGED SCALE.

while the intensity of the radiation far exceeded anything practicable in the preparation of ordinary commercial radium compounds.

The authors have made systematic tests to determine as accurately as possible the loss of brilliancy of the radium compound ordinarily used, both in powder form, before painting,

\* *Loc. cit.*, 216.

and also after mixture with the varnish used in its application to the surface to be treated.

(a) *Luminous Compounds Before Painting.*—Nine samples from several makers, and containing different proportions of radium salt mixed with various qualities of zinc sulphide have been retained under observation at the National Physical Laboratory. In some cases it has been possible to obtain measurements of the brightness at intervals over a period of

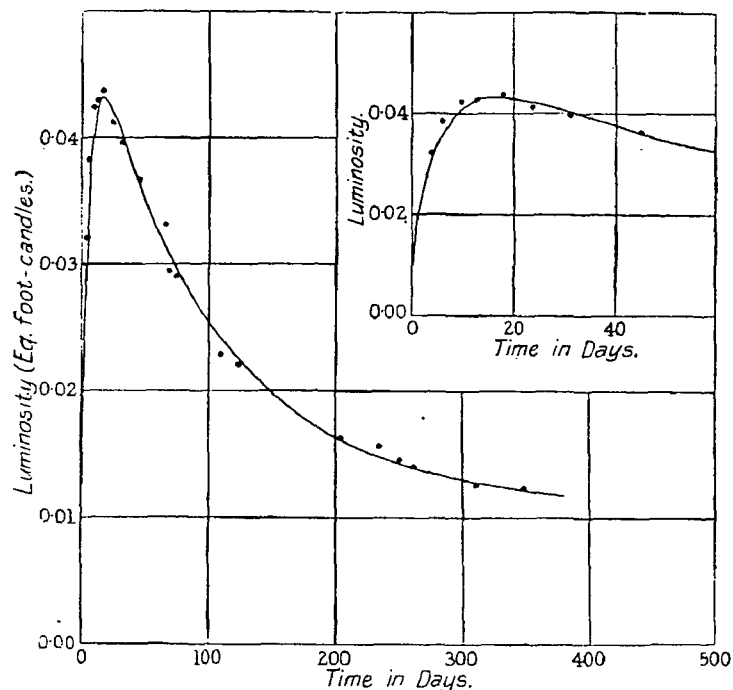


FIG. 8.—DIAGRAM SHOWING THE VARIATION THROUGHOUT LIFE OF THE LUMINOSITY OF SAMPLE 8 OF RADIUM LUMINOUS COMPOUND. INSET, THE EARLY PORTION OF THE LIFE ON AN ENLARGED SCALE.

about 15 months, and curves showing the rise and fall with time of the luminosity of two characteristic samples are shown in Figs. 7 and 8.

In samples 3 and 8 (Figs. 7 and 8) measurements were possible within a short time after mixing the radium salt with the zinc sulphide. The curves referred to above have an inset showing in greater detail the earlier history of these samples. It is seen that there is a growth of luminosity to a peak value which is attained generally about 15 days after mixing. The

characteristics of the early life of luminous compound are of special interest as bearing on the theory underlying the action of radium on zinc sulphide.

The values of luminosity observed during the first 40 days are taken from the curves and scheduled in the following table:—

TABLE IV.

Period after mixing, Days	Luminosity (equivalent foot-candles).			
	Sample 3.		Sample 8.	
	Actual.	P.c. of maximum.	Actual.	P.c. of maximum.
4	0.032	71	0.030	70
6	0.038	84	0.035	81
8	0.041	91	0.039	91
10	0.043	95	0.041	95
15	0.045	100	0.043	100
20	0.045	100	0.043	100
25	0.044	98	0.042	98
30	0.043	96	0.040	93
35	0.042	93	0.039	91
40	0.041	91	0.037	86

It should be noted that these samples were from different makers, the method of mixing the radium salt with the zinc sulphide being stated to be different in the two cases. In the first sample the mixture was made with both salts in the dry state, while in the second the radium salt was added in the form of a solution to the zinc sulphide, the resulting mixture being then evaporated to dryness. With this latter method of mixing it is certain that nearly all the radium emanation is driven off, so that the subsequent increase of luminosity of the radium paint is due to the gradual growth of the radium emanation and its products radium A and radium C.

After attaining its maximum value at the end of about 10 to 20 days from mixing, the luminosity then begins to decrease, at first very gradually and then more rapidly until, after a period of about six to seven weeks from the date of mixing, the rate of decay appears to have settled down in accordance with an exponential law—i.e., the rate of decay ( $k$ ) of luminosity ( $B$ ) at any time is approximately proportional to the value of the luminosity at that time and  $B/B_0 = e^{-kt}$ . The logarithm of the luminosity therefore plotted against time should give a straight line. The logarithmic curve corresponding with the results of sample 3 (Fig. 7) is shown in Fig. 9. After

an initial period of 40 days it will be seen that the logarithmic relation given above is nearly fulfilled up to the end of about 150 to 200 days, but that after this period the luminosity does not diminish so rapidly as would be expected from the above law. It will be shown in a further Paper \* that the effect may be explained on the assumption that superposed on the normal

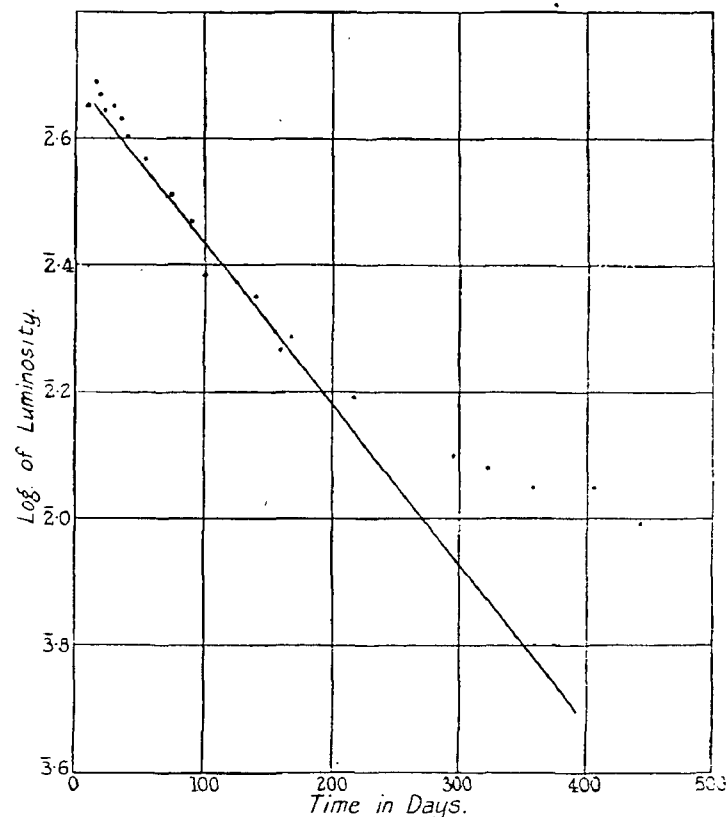


FIG. 9.—DIAGRAM SHOWING THE GRADUAL DEPARTURE OF THE OBSERVED VALUES OF LUMINOSITY OF SAMPLE 3 FROM THE LOGARITHMIC RELATIONSHIP.

decay proportional to the luminosity, there is a smaller effect operating in the reverse direction, which becomes relatively more and more important as the luminosity becomes smaller. The observations have not continued long enough to enable the relative importance of these effects to be determined with great precision, but the results point to the conclusion that after

\* "The Theory of Decay in Radioactive Luminous Compounds," by J. W. T. Walsh.

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about 500 days the luminosity will attain a value which is sensibly constant. This period, however, is not the same for all compounds, as in some of them the rate of decay appears to diminish more rapidly than in others. The effect on the rate of decay of the amount of radium present is shown by the curves in Figs. 10 and 11. Each of the two pairs of compounds 1

Days.

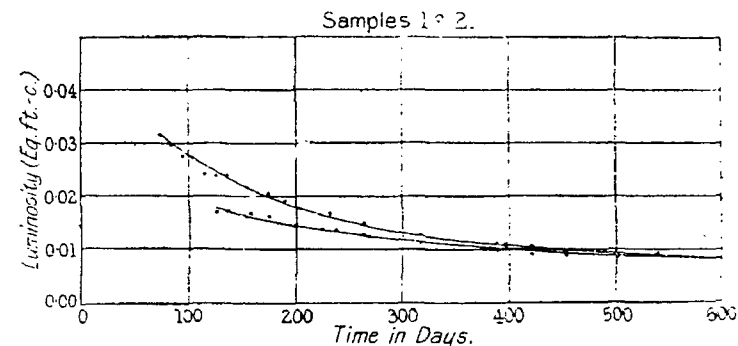


FIG. 10.—LUMINOSITY CURVES FOR SAMPLES 1 AND 2 OF DIFFERENT RADIUM CONTENTS, SHOWING THE SLOWER RATE OF DECAY OF COMPOUND CONTAINING HALF QUANTITY OF RADIUM.

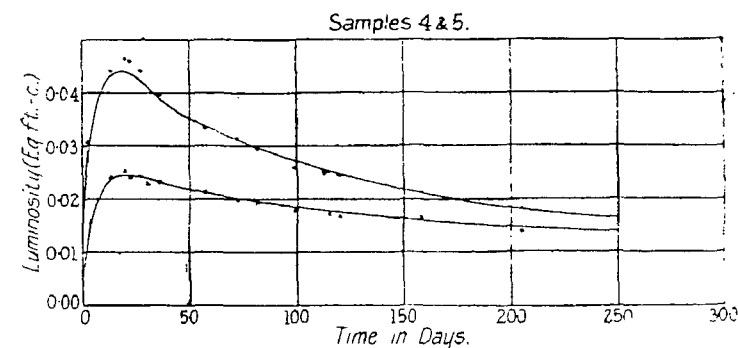


FIG. 11.—THE EARLY PORTIONS OF THE LUMINOSITY CURVES FOR SAMPLES 4 AND 5 OF COMPOUND CONTAINING RADIUM IN THE PROPORTION OF 2:1

and 2 (Fig. 10) and 4 and 5 (Fig. 11) was made up at the same time by the same manufacturer with an identical quality of zinc sulphide. One sample of each pair was prepared with exactly half the quantity of radium salt contained in the other. In the case of the first pair, observations were not made until about 100 days after mixing, while in the case of the second pair the first five months' observations are all that have so far

been obtained. It will be seen that the initial brightness is in the ratio of the radium content, but the rate of decay of the compound of smaller content is slower, so that the ratio of the luminosities gradually approaches unity as time goes on. This question is one of considerable commercial importance, for it is clear that if the compound is required for use after the lapse of several years, there is very little advantage, except from the point of view of initial brightness, in increasing the radium content beyond a certain point. From present

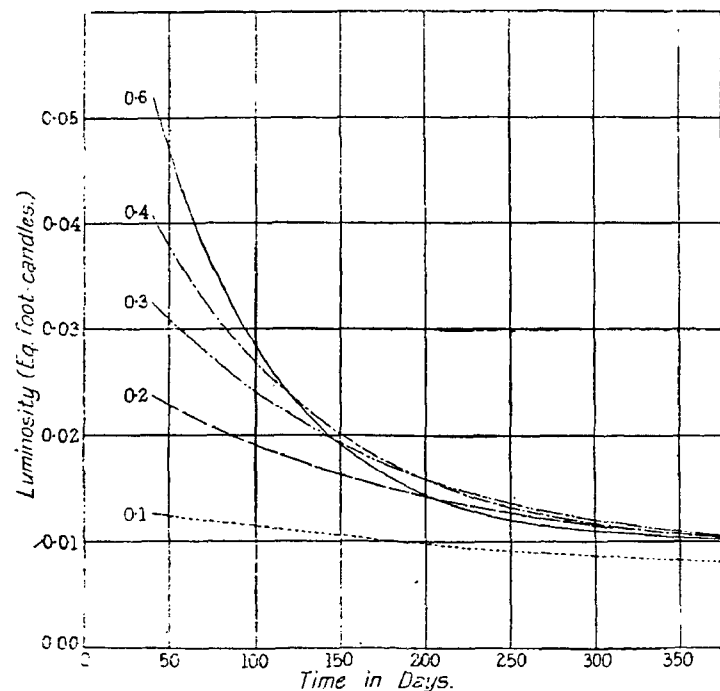


FIG. 12.—THEORETICAL LUMINOSITY CURVES FOR SAMPLES OF RADIUM CONTENT (MG./GM.) SHOWN AGAINST EACH CURVE. (CONSTANTS OF DECAY ASSUMED OF FAIR AVERAGE VALUES.)

knowledge it would appear that for short period use (say three to four months) the radium content may be usefully as high as 0.4 mg. per gm. Fig. 12 shows a family of luminosity curves based on the average data at present obtained for compounds made up with different proportions of radium salt in the same quality of zinc sulphide, this sulphide being assumed to have characteristic constants of a fair average value.

It is to be noted that in this diagram in which the effect of

the diminution of decay with time is allowed for, the curves for 0.4 and 0.2 mg. compound do not cross one another as would curves based on a purely logarithmic law with constant rate of decay throughout.

(b) *Painted Dials*.—Seven dials painted with different samples of 0.4 mg. luminous compound have been kept for the past 12 months, and their luminosities have been found to decay to approximately half value in that period. The actual results of the measurements are given in Table IV. The dials are all ordinary metal dials used for aeroplane instruments, painted with a matt black surface, and having the luminous compound laid on in fine lines about 0.2 mm. to 0.3 mm. high to mark the graduations and figures of the dial. These dials were tested in the apparatus described on p. 229, with the following results:—

TABLE V.

Dial No.	Initial luminosity (p.c. of standard).	Luminosity after 11 months (p.c. of standard).	Decay of luminosity (p.c. of initial luminosity).
1,406	94	60	63
995	82	46	51
1,470	94	58	62
1,783	63	34	54
1,794	24	(16)	(67)
1,496	91	40	44
1,497	52	25	48
Average .....			55 p.c.

It is to be remarked that the rate of decay of luminosity of the dials is noticeably lower than that of the compound before application, and as has been said, the initial luminosity of the painted dial is also smaller, being of the order of one-third to one-fourth of that of the compound before application. The reason for this is not clear, but it seems reasonable to suppose that the effect of the varnish or other medium is to impede the activity of the  $\alpha$ -ray bombardment from the radium and in this way to cause an effective lowering of the radium content of the compound with a subsequent reduction of both the initial luminosity and also the rate of decay. It is not suggested that the whole effect is due to such a cause, but it seems likely that some such effect as this is operative to account for at least a portion of the observed effect. It is significant, for instance, that the luminosity standards for the dry compound and for the painted dials are 0.03 and 0.0075 ft.-candles respectively

(i.e., in the ratio of 4:1), while, if we assume curve 0.4 of Fig. 12 to represent the behaviour of the compound before application to dials, the relative behaviour *after* application would be very well represented, both as regards initial brightness and rate of decay, by a curve lying between those for 0.2 and 0.1 of the same diagram.

#### 7. Considerations Governing the Choice and Specification of Radium Luminous Compound.

(a) *Radium Content.*—It will be clear from what has been stated above concerning the relation between the radium content of a compound and the rate of decay of its luminosity, that, in specifying a standard for any particular purpose, the probable life of the compound in actual use must be the principal deciding factor as to the particular radium content to be employed. The factor of cost, however, must not be ignored. Compound containing 0.4 mg. of radium per gramme of zinc sulphide costs about £6 to £7 per gramme. This is mainly due to the amount of radium present, and the cost may, therefore, be considered as roughly proportional to radium content.

Further, from what has been said above (p. 239) as to the relative rates of decay of radium compound in powder form and after application to dials, it will be seen that in considering the standards to be adopted in the two cases, the radium content of compound *after application* must be considered as reduced to about one-third, in order to ascertain its rate of decay from the data given in the diagram of Fig. 12, p. 238, which refer to the powder only; that is to say, a *dial* painted with compound containing 0.3 mg. per gram. of radium will follow approximately the same decay curve as *powder* containing 0.1 mg. per gram.

For the compound alone (as distinct from painted dials) after the first six months, the luminosities of compounds of all contents between 0.2 mg. per gramme and 0.8 mg. per gramme are the same to within 20 per cent.

Considering, first, the upper limits desirable for radium content, it is seen that if the compound is likely to be required for more than six months, the use of a compound of radium content greater than 0.2 mg. per gramme can only be justified by a need for greater brilliancy during the early part of the life.

Similarly, it may be said that for painted dials the maximum useful radium content for a life of over six months will not be more than 0.6 mg. per gramme.

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In the case of instruments of which the life under normal working conditions is appreciably less than six months the use of a compound containing, say, 0.6 mg. per gramme or over might be of advantage, provided it could be ensured (a) that the dials were painted within three weeks of the preparation of the compound, and (b) that the instruments were not kept in store for more than a month after painting. Probably, in ordinary circumstances, these conditions would not be easy of realisation, and the use, therefore, of a compound of greater radium content than about 0.4 mg. per gramme may be ruled out. Even for 0.4 mg. compound it is important that the interval between the preparation of the compound and its application to the dial should be as short as possible, for it is clear that deterioration in luminosity takes place from twice to three times as rapidly in the powder as in the painted dial. Thus, a deterioration due to one month's delay in painting will be as much as that resulting from about three months' storage of the completed dial.

Referring next to the lower limits desirable for radium content, we see that for cases in which the compound is employed in powder form, the minimum useful radium content is only a little below 0.2 mg. per gramme, and except on the score of expense alone it is not desirable to employ a compound of less radium content than this, because the luminosity of the 0.1 mg. compound after an indefinitely long period, is still nearly 20 per cent. below that of the 0.2 compound. Hence, even in cases where very long life is desirable without the possibility of replacement, the use of the higher grade material is to be preferred unless the cost of the radium compound used has a preponderating influence on the price of the complete instrument. In such cases, rather less than half the cost would be saved and 20 per cent. of the final luminosity would be sacrificed by the adoption of 0.1 mg. per gramme for the compound in powder form.

Similarly for compound used in painting dials, 0.4 mg. per gramme is the best content to use if final luminosity is not to be sacrificed. Some 50 per cent. saving of cost should result from the use of compound for painting dials containing 0.2 mg. per gramme, but the final luminosity would be somewhat less. It should be remembered that there is nothing appreciable to be gained by the use of compounds for dials *below* this last named limit because it will be nearly as economical, and a better result will be achieved by employing a smaller quantity

of the better grades of compound instead of the large quantity of a compound of lower radium content.

(b) *Luminosity*.—The standard of luminosity for the luminous compound in powder form should be so fixed as to enable a good grade of compound to pass the test within a reasonable period after its preparation. The present standard for 0.4 mg. compound (*viz.*, 0.03<sub>0</sub> f.c.) is easily attainable in a good grade three months after preparation. This time limit may be extended for grades of lower content to, say, four months for the 0.2 mg. compound and six months for 0.1 compound. The corresponding luminosity standards for the three grades 0.4, 0.2 and 0.1 are then 100, 60 and 35 per cent. of the present National Physical Laboratory standard. For painted dials the standards of luminosity should be 25 per cent. of the standards fixed for the powders from which they are to be painted. The above-named figures of 100, 60 and 35 per cent. will then express the standards of the painted dials in terms of the present National Physical Laboratory standard for dials—*i.e.*, 0.0075 equivalent ft.-candles.

The above conclusions have been based on the results obtained by observation of the samples of paint at the National Physical Laboratory from October, 1915, to January, 1917. None of these compounds have a radium content greater than 0.4 mg. per gramme, so that the behaviour of such grades of compound can only be predicted on the assumption that they will obey the same laws as those which have been found to hold for the samples of lower radium content. Further, a series of observations extending over several years would be necessary for the certain prediction of the behaviour of compound for long periods. It has been necessary, however, to make decisions from the results already obtained and the laws deduced from them, without the long delay necessary for experimental confirmation. The conclusions given above are those which experiment has so far shown to be most likely to be justified.

These conclusions are briefly summarised in the two following tables.

CASE A.—Compound Used in Powder Form.

Period of enhanced luminosity.	Radium* content.	Luminosity standard.
About six months ...	0.3 mg./gm.	80 p.c. of N.P.L. standard for compound
About one year ...	0.2 mg./gm.	60 p.c. " " "

\* Expressed in gamma ray activity of an equivalent weight of hydrated radium bromide.

CASE B.—Compound Used for Painting Dials.

Period of enhanced luminosity.	Radium content.	Luminosity standard.
About six months ...	0.6 mg./gm.	140 p.c. of N.P.L. standard for compound
About one year ...	0.4 mg./gm.	100 p.c. " "
Sensibly uniform luminosity with a final value about 30 p.c. below that of the other grades	0.2 mg./gm.	60 p.c. " "

The authors desire to express their acknowledgments to Capt. Creagh-Osborne, Admiralty Superintendent of Compasses, and his assistant, Lieut. Bray. They also wish to express their warm appreciation of the co-operation of manufacturers and others in the work which has been done, and particularly that of Mr. F. H. Glew and Mr. H. A. Hughes.

## APPENDIX A.

### ABSORPTION OF $\gamma$ -RAYS BY THE ZINC SULPHIDE IN RADIUM LUMINOUS COMPOUND.

(a) *Approximate Graphical Method*.—The first step is to find the difference between the emergent intensities for axial con-

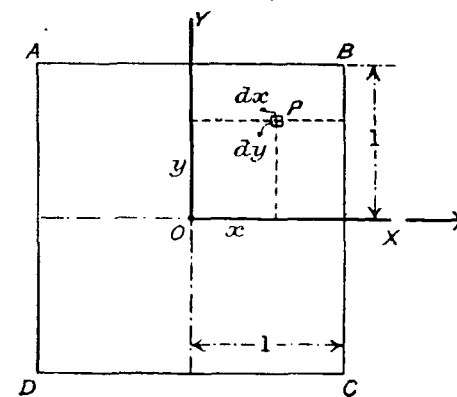


FIG. 13.

centration and for uniform distribution of radium in the case of a vessel of square section.

Let  $ABCD$  be a section of the vessel of side  $2l$ ,  $O$  being the

axis. Then, for axial concentration the intensity in the direction of the  $OX$  axis is

$$Ie^{-\lambda l} = I \left\{ 1 - \lambda l + \frac{\lambda^2 l^2}{2!} - \dots \right\}$$

For uniform concentration, however, consider the elementary particle  $P$ . The intensity due to this particle  $P$  of area  $dxdy$  is

$$\frac{dxdy}{4l^2} \cdot I \cdot e^{-\lambda(l-x)}.$$

Hence, the total intensity for the whole section is—

$$\begin{aligned} & \left( \frac{I}{4l^2} \right) \int_{-l}^{+l} \int_{-l}^{+l} e^{-\lambda(l-x)} dxdy \\ &= \frac{I}{2\lambda l} (1 - e^{-2\lambda l}), \\ &= I \left\{ 1 - \lambda l + \frac{\lambda^2 l^2}{2!} - \dots \right\} \end{aligned}$$

It will be seen that the two expressions obtained above are equal to 0.1 per cent. if  $\lambda l < 0.1$ , while if  $\lambda l < 0.01$  the expressions are identical to 10 parts in a million.

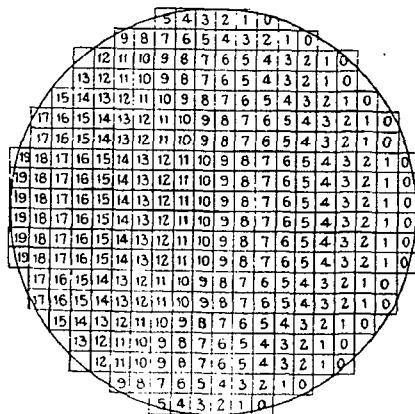


FIG. 14.

Now,  $\lambda$  has been experimentally determined by placing a small tube of radium axially in a cylindrical vessel of zinc sulphide, of which the diameter was 24 mm. The intensity thus found was 0.895  $I$ , so that

$$e^{-\lambda l} = 0.895,$$

giving

$$\lambda = 0.00025 \text{ mm}^{-1}.$$

Now, consider the case of a cylinder of radius  $a$ , and let this

be divided into a number of small vessels of square section of side  $2b = a/10$ , as shown in Fig. 14.

It is clear that in this manner uniform distribution over the cylinder may be considered as equivalent to that due to concentration at the axes of 156 square vessels of side  $a/10$ . Thus the intensity in the direction of the arrow due to each vessel marked "0" will be  $(I/312)e^{-\lambda b}$ , that due to each vessel marked "1" will be  $(I/312)e^{-3\lambda b}$ , and so on. In this way it is found that for a vessel of 24 mm. diameter, the intensity is (0.910)  $I$ , instead of the (0.895)  $I$ , due to axial concentration.

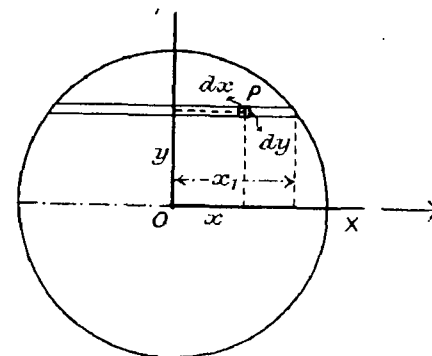


FIG. 15.

(b) *Rigid Mathematical Determination.*—Let  $O$  be the axis of a cylindrical vessel of radius  $a$ , and let  $P$  be an elementary particle of area  $dxdy$ . Then the intensity along the  $OX$  axis due to the particle at  $P$  will be

$$\frac{I}{\pi a^2} dxdy e^{-\lambda[\sqrt{a^2 - y^2} - x]}.$$

Hence, the total intensity is—

$$\begin{aligned} I' &= \frac{I}{\pi a^2} \int_{y=-a}^{y=+a} \int_{x=-\sqrt{a^2 - y^2}}^{x=+\sqrt{a^2 - y^2}} e^{-\lambda[\sqrt{a^2 - y^2} - x]} dxdy, \\ &= \frac{2I}{\lambda \pi a^2} \int_0^a (1 - e^{-2\lambda \sqrt{a^2 - y^2}}) dy. \end{aligned}$$

Expanding the exponential and integrating each term this gives—

$$\begin{aligned} I - \frac{\lambda}{a} \cdot \frac{\pi a^2 I'}{2I} &= 1 - 2\lambda a \cdot \frac{\pi}{4} + \frac{4\lambda^2 a^2}{2!} \cdot \frac{2}{3} - \dots \\ &+ (-1)^r \frac{(\lambda a)^r}{r!} \cdot \frac{\pi}{2} \cdot \frac{\Gamma(r+2)/2!}{\Gamma(r+3)/2!} + \dots \end{aligned}$$



This gives—

$$I'/I = 1 - \frac{8}{3\pi}(\lambda a) + \frac{1}{2}(\lambda^2 a^2) - \frac{32}{45\pi}(\lambda^3 a^3) + \dots$$

It will be seen that this series is rapidly converging for all values of  $\lambda a$  up to unity, and for the sake of convenience the values of the first 12 coefficients are tabulated below. These will enable the value of  $I'/I$  to be calculated correct to 0.01 per cent. for all values of  $\lambda a$  up to unity.

Table.

The first 12 coefficients in the expression for the  $\gamma$ -ray activity of a sample of radium compound contained in a cylindrical vessel, viz. :—

$$I'/I = z_0 + z_1(\lambda a) + z_2(\lambda^2 a^2) + \dots$$

Coefficient.	Value.	Coefficient.	Value.
$Z_0$	+1.000000	$Z_7$	-0.001642
$Z_1$	-0.848826	$Z_8$	+0.000347
$Z_2$	+0.500000	$Z_9$	-0.000067
$Z_3$	-0.226354	$Z_{10}$	+0.000012
$Z_4$	+0.083333	$Z_{11}$	-0.000002
$Z_5$	-0.025869	$Z_{12}$	+0.000000
$Z_6$	+0.006944		

Assuming the value for  $\lambda$  given above  $\lambda a = 0.111$  for  $a = 12$ , so that  $I'/I = 0.9116$ , with which the value 0.910 found by the graphical method is in good agreement.

#### ABSTRACT.

The Paper contains the results of measurements made on various samples of radium luminous compound during the last two years. Determinations of the brightness of the compound in powder form and when made up into paint, and also after the application of the paint to instrument dials, were carried out; and curves are given showing the rates of decay of luminosity. The radium contents of the compounds were determined by comparison of their  $\gamma$ -ray activities with that of a preparation of pure radium bromide, which is periodically compared with the British radium standard. The various precautions which have to be observed, and the corrections which have to be applied in making the various determinations are explained, and the considerations which should govern the proportion of radium employed for practical purposes are discussed.

#### DISCUSSION.

Mr. F. H. GLEW. The difference between some of Marsden's results and those of Dewar and Crookes had been attributed to some difference in the zinc sulphide employed. He could state that the sulphide supplied to the National Physical Laboratory was of different origin from either of these. Before the war we were almost entirely dependent on foreign sources for the supply of these materials, but we are now quite self-supporting. In

Marsden's work the inside of the tube was completely coated with sulphide, so that the effects were observed by light transmitted through the material. It was better to paint only one half of the tube so that the inner surface, which was exposed to the bombardment, could be directly observed. In the photometer measurements, was there any advantage in placing the coloured filter between the light and the reflecting surface? He preferred to have it in the eye-piece. Earlier records of the coloration due to absorption would be of value. In testing sulphide for  $\gamma$ -ray activity he placed it in contact with the ionisation chamber. Less material was then required for the test, which was an advantage. The authors state only the diameters of the tubes employed; but he had found that there were differences in the results obtained with tubes horizontal or vertical. The length of the tube, therefore, appeared to be an important factor. Further, the results given by a small quantity of sulphide in a large tube would be too high; for the space above the sulphide is filled with the emanation gas, and the  $\gamma$  radiation from it is not subject to the absorption of the sulphide, as is the radiation from the latter itself. The discrepancy between the observed and calculated values of the absorption was probably important. Was this due to some selective action, the sulphide transforming the  $\gamma$  radiation into radiation of some other type? It was singular that specimens prepared by the dry and wet methods should be so similar in their properties after only 4 days; for in the dry method the emanation and all its products are retained.

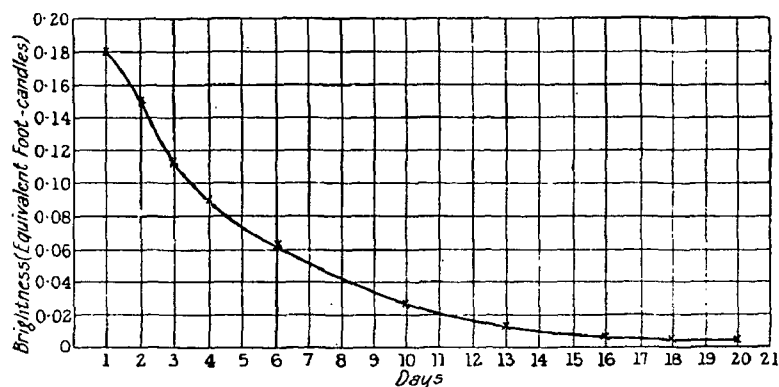
Dr. MAKOWER stated that by using an electroscope of small capacity good  $\gamma$ -ray measurements could be obtained with much less than 1 milligram of radium.

Dr. H. S. ALLEN asked if the zinc sulphide employed was wholly crystalline or partially amorphous. An investigation of its physical condition would be useful.

Prof. CLINTON said that the difficulties of these measurements were very great and the authors were to be congratulated on having overcome them so effectively.

Mr. J. S. DOW said the Paper contained a great deal of useful information. It was common knowledge that a great deal of work on these luminous paints was being done, and they were fortunate in having these results made public. No doubt after the war even fuller information would be forthcoming. His own experience was generally in accord with a number of important results announced by the authors. For example, he found that freshly-prepared compound of 0.4 composition had a brightness not far removed from that mentioned. He rather doubted whether the absolute brightness could be given with exactitude, though an empirical result could no doubt be reproduced quite closely in the laboratory. He had noticed, for example, that if two patches of zinc sulphide and calcium sulphide were excited by visible light, their brightness might be equal when viewed at a distance of a few inches; but the zinc sulphide (giving a green light) appeared much the brighter as the distance of the observer is increased. There was also the question whether the colour-match obtained by using a blue-green glass with an incandescent lamp was sufficiently close to eliminate the purkinje effect at low illumination. With low luminosity materials he had found that a slight obliquity of gaze affected the comparison. He agreed that the colour required was a green with a distinctly blue component. It was curious, however, that while with bright specimens any colour departure was evident almost any green glass appeared to give a colour match at weak luminosities. He had also found that the colour of freshly-prepared and very bright samples was distinctly more blue in tint than that of old and weak ones. The authors' suggestion that the most satisfactory compound, from the standpoint of luminosity in the later period of life and minimum cost, was the 0.2% material, was a most important result. He thought this would probably be borne out by later experience. In view of the expense, it was

obviously necessary to make the radium content as small as possible. Another point on which little information was forthcoming was the order of luminosity necessary in practice for different purposes. Such consideration would probably also favour the use of 0.2 material in most instances. In his experience, in complete darkness, much brighter material might actually cause irradiation. He would like to ask whether the authors had found that in the weaker compounds the initial rise in brightness was disproportionately great and more prolonged than would be expected from the brighter specimens. This was a feature of some tests of which he had received particulars from the United States, and was in some measure supported by his own experiments. Finally, it might be of interest to refer to a series of observations he had recently made of the brightness of a tube coated internally with zinc sulphide and filled with radium emanation, which Mr. Glew had kindly placed at his disposal for test. The initial brightness of this tube



DECAY OF LUMINOSITY OF TUBE FILLED WITH RADIUM EMANATION AND COATED INTERNALLY WITH ZINC SULPHIDE IN THE COURSE OF THREE WEEKS AFTER PREPARATION.

was relatively high—of the order of 0.2 foot-candles; but in three weeks it fell to 1/60th of its initial value. Naturally such a tube would decay much more rapidly than a compound containing powder. Another feature was the very clear exhibition of the formation of local patches of brightness due to condensation of the emanation, to which Mr. Glew had referred. Patches, constantly changing their position, as bright as four times the value of the main luminosity would make their appearance, and then fade away and reappear in another part of the tube. This effect was marked in the early stages of the life, but much less evident at a later stage, when the general brightness had diminished. It would be interesting to know if such effects were met with in varnish or powder materials. In his personal experience he had met only one case, an exceptionally bright tube which was initially much brighter at one end than the other. A few days after the first test the inequality disappeared and had not been noted since.

Dr. LEVY said he had found the luminosity of different samples of zinc sulphide to vary perceptibly in tint when stimulated by daylight, but had not noticed this with  $\alpha$ -ray stimulation. In his experience, if a sample of sulphide gave good results when stimulated by daylight, it would also be good when stimulated by radium. The reverse did not hold, however, as a specimen might be poor with daylight, but good with radium excitation.

Mr. T. SMITH thought the coloured filter used in the photometric measurements should have been placed as near the lamp as possible,

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since such a filter acts, to a certain extent, as a self-luminous source. If the tube containing the compound was not much wider than the aperture in the screen, a considerable loss of light would occur, near the edges of the aperture, due to oblique internal reflection at the walls of the tube. For accurate work it was important to have a uniformly illuminated field, and he suggested the desirability of employing tubes with a flat side. Where the tubes were too small to fill the aperture satisfactorily, an optical system could be used to give an enlarged image of the tube and so avoid refractions at large angles. The absorption of the system could be found experimentally with a larger tube.

Dr. A. RUSSELL laid stress on the practical importance of the Paper. He had at times been asked by manufacturers to measure the visibility of watch faces in the dark. The problem was so difficult that hitherto he had done nothing. The present Paper suggested the required practical tests, and a first draft of a specification for securing a standard material could now be written. Progress would be delayed if they waited till all the associated physical and physiological problems were cleared up.

Dr. S. RUSS (in a communication which was read by the Secretary) said the decay curves differed appreciably from those of Marsden for Willemite under intense  $\alpha$ -ray bombardment. Did the theory held by the authors as to the mechanism of the action differ from that advanced by Prof. Rutherford to explain Marsden's results? Had the authors any measurements of the rate of decay of efficiency of fluorescent screens under the action of X-rays?

Mr. WALSH, in reply, said that if the coloured filter were employed in front of the eye, as suggested by Mr. Glew, its coefficient of transmission for the light emitted by the compound would require to be determined and introduced in the results as well as the coefficient for white light. This would add to the uncertainties of the determinations. To detect change of colour with age would have been difficult in their experiments because of the continuous decrease in luminosity. They had detected differences in colour in samples sent by different makers; but were unaware whether these were from the same source, or made in the same way. The authors had no data on the relation between daylight and radium excitation, and Dr. Levy's remarks were of great interest. The order of accuracy (about one per cent.) was not found to be appreciably affected by altering the position of the coloured screen. The diameter of the smallest tubes usually employed was not less than about twice the apparent width of the aperture. It was not convenient to use a smaller aperture, and they had to do their best with the tubes in which the material was sent. They had not found any samples of radium compound which gave the slow initial rise of luminosity mentioned by Mr. Dow. It is possible, however, that if some radio-active agent other than radium were employed, such, for example, as mesothorium, a slow rise of luminosity might very likely result. They would be interested to have further details of the samples referred to by Mr. Dow in this connection.

Mr. HIGGINS, replying to Dr. Makower, said that they had to employ the radium standards which were available, and these were not suitable for use with a small capacity electroscop. In reply to Mr. Glew, the tube should not be close to the electroscop. If it was 30 to 40 cm. off, the orientation of the tube was of little importance, and small corrections could be applied. The errors introduced by having it too close were similar in character to those produced if an extended source of light is used too near a photometer head. They suspected selective action to be at the root of the discrepancy in the absorptions, but could not definitely say. To investigate the physical properties of the sulphide would be a very long task, and they had not attempted it in the present research.